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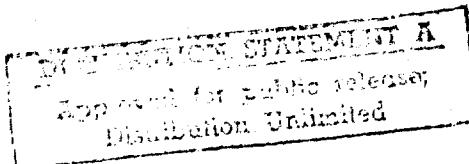
INFRARED CONTINUUM ABSORPTION BY ATMOSPHERIC WATER VAPOR IN THE 8-12 μ m WINDOW

Robert E. Roberts
Lucien M. Biberman
John E. A. Selby

April 1976



INSTITUTE FOR DEFENSE ANALYSES
SCIENCE AND TECHNOLOGY DIVISION



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continuum absorption coefficient and the elimination of the atmospheric broadened continuum term. Finally, and most critically, a strong measured temperature dependence must be included in the water vapor continuum absorption coefficient. For path lengths ranging from 10 to 50 km, failure to incorporate these corrections can lead to errors in the computed transmission ranging from factors of 2 to more than 10,000.

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This analysis has been part of a larger analysis of the effect of weather upon infrared propagation being carried out under the IDA Central Research Program.

ABSTRACT

We have carried out a detailed analysis of several long-path-length transmission measurements in the 8 to 12 μm atmospheric window in order to determine the extinction coefficient due to the water vapor continuum. Our results indicate that three modifications to the current LOWTRAN atmospheric transmission model are required. The first two corrections are an improved fit to the pure water vapor continuum absorption coefficient and the elimination of the atmospheric broadened continuum term. Finally, and most critically, a strong measured temperature dependence must be included in the water vapor continuum absorption coefficient. For path lengths ranging from 10 to 50 km, failure to incorporate these corrections can lead to errors in the computed transmission ranging from factors of 2 to more than 10,000.

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I. INTRODUCTION

A valid atmospheric transmission model for infrared (IR) radiation is of critical importance to (a) the selection of optimal spectral bands for IR imaging systems, (b) the evaluation of sensor performance under varying weather conditions, and (c) the calculation of energy fluxes in planetary atmospheres for the study of climatology (Refs. 1-3). Of the many models available for the calculation of atmospheric transmission, the LOWTRAN model of Selby and McFlathey (Ref. 4) is probably the most used and useful. It, however, has been the object of some concern since several comparisons (Ref. 5) of LOWTRAN predictions with recent measurements in the 8-12 μm window indicate that LOWTRAN transmission predictions are usually low (often by a factor of two or more for long atmospheric paths). In an effort to verify such a discrepancy, and if possible to correct it, arrangements were made to obtain the extensive long-path-length transmission measurements of EMI (Ref. 6) in order to undertake a careful analysis of their raw data, since it provided a nearly continuous record of transmission in multiple bands together with the meteorological conditions. This data facilitated a separation of effects due to gaseous and aerosol atmospheric components and thus provided a means for evaluating some of the factors of the LOWTRAN model. In this spectral region the LOWTRAN model is currently based on the measurements of Burch (Ref. 7) and McCoy et al. (Ref. 8). Using the EMI data base together with other laboratory measurements, we will address the problem of the water vapor continuum and its effect upon the transmission

in the 8-12 μm band.

The amount of radiant energy transferred by the atmosphere is determined by two principle types of constituents, namely gaseous molecules and aerosols or particulates with their respective extinction coefficients σ_{mol} and σ_{aer} . At a particular wavelength the transmission is given by*

$$T = \exp(-\sigma_{\text{tot}} L) \quad (1)$$

where the total extinction coefficient is $\sigma_{\text{tot}} = \sigma_{\text{mol}} + \sigma_{\text{aer}}$ and L represents the path length.** The magnitude of σ_{mol} or σ_{aer} clearly depends upon the optical properties, atmospheric concentration, and temperature of the molecular or particulate species.

In this paper our primary interest is gaseous absorption, particularly the contribution due to water vapor. Furthermore, we will restrict our attention to the 8-12 μm window region where the so-called water continuum absorption tends to play a dominant role.*** Most of the broadband molecular absorption is due to CO_2 and H_2O .[†] The absorption by CO_2 is the best understood of the two atmospheric components and can be estimated accurately with the current LOWTRAN program (Ref. 4) provided its concentration is known. In the 8-12 μm region the extinction by CO_2 is typically a small part of the total, i.e., $\sigma_{\text{CO}_2} \approx 0.02 \text{ km}^{-1}$, whereas $\sigma_{\text{tot}} \gtrsim 0.15 \text{ km}^{-1}$. The dominant contribution to the total extinction coefficient and the "weak

* For broadband measurements the extinction coefficients are defined by the appropriate bandwidth-average transmission.

** In this paper σ_{tot} will be given in km^{-1} and L in km.

*** In our analyses and fit of the 8-12 μm window we will include continuum data out to 30 μm .

[†] There is also some narrow line absorption due to O_3 in this region, but it is not important for sea-level or broadband measurements as discussed here.

link" in the current atmospheric models is the part due to water vapor, σ_{H_2O} , and aerosols, σ_{aer} . Our immediate interest here is to review and analyze selected transmission data with the intent of isolating and improving the estimate of the water vapor component. Although the relative contribution by aerosols is subject to large fluctuations depending on the presence or absence of fogs, dust, sea spray, etc., the contribution by water vapor is omnipresent but has minimum effects in winter climates where the partial pressure of water, P_{H_2O} , is small. For a typical range (Ref. 6) of mid-latitude vapor pressures, 4 torr $\leq P_{H_2O} \leq 14$ torr, the water vapor extinction varies from 0.1 to 0.4 km^{-1} over the band 7.9-11.3 μm . On the other hand, the aerosol extinction is essentially zero for extremely clear days (visual ranges $\gtrsim 30 \text{ km}$) and can become larger than 1 km^{-1} for foggy, low-visibility conditions. The analysis presented here will be confined to the EMI atmospheric transmission measurements (Ref. 6) made under conditions of extremely high visual ranges ($\gtrsim 38 \text{ km}$) or to laboratory-controlled experiments (Refs. 7-10); hence, the aerosol contribution is minimal.*

*For the present analysis it is not at all necessary for σ_{aer} (or σ_{CO_2}) to be identically zero. The important point is that it be a constant, independent of P_{H_2O} .

II. THE EMI DATA BASE

We have used two sources of data for our analysis of the H_2O continuum absorption, the first being the EMI (Ref. 6) measurements of transmission for a 20-km path over open salt water near Cornwall, off the coast of England.* The second represents a critical review of 8-12 μm laboratory measurements (Refs. 7-10).

EMI reports 103 sets of unobscured measurements with visibilities of at least 38 km. Each set includes seven bands from 0.57 to 11.3 μm , including the laser lines at 0.63 μm (HeNe) and 10.59 μm (CO_2). We have chosen to analyze their 7.9 to 11.3 μm and 10.59 μm data, since it represents the region where the H_2O vapor continuum absorption plays a dominant role. The partial pressure of water vapor for their high visibility measurements varies from 4 to 14 torr with a small mean seasonal temperature variation of $\pm 4K$.** We have analyzed the 7.9 to 11.3 μm band with respect to the water vapor pressures, P_{H_2O} , according to three data processing methods,*** and then compared the P_{H_2O} dependence with what is predicted by the current LOWTRAN

* Our analysis of the EMI measurements was actually based upon the complete computer data tapes rather than the summaries represented by the EMI report (Ref. 6).

** The summer and winter means were 16C and 8C, respectively.

*** The methods employed include (a) linear filtering combined with linear regression on the filtered data, (b) straightforward linear regression on the entire 103-point data base, and (c) linear regression on the 15 "best" transmission measurements for each absolute humidity; the best data were simply chosen by selecting the lowest extinction coefficient for each P_{H_2O} .

routine (Ref. 4). The LOWTRAN transmission model includes two contributions to σ_{H_2O} : one due to molecular line absorption $\sigma_{H_2O \text{ mol}}$, which is relatively well understood and predictable, and the other so-called continuum absorption, $\sigma_{H_2O \text{ cont}}$, which is not at all well understood and furthermore dominates the total water vapor absorption σ_{H_2O} .* Various mechanisms have been proposed to explain the water vapor continuum absorption, including contributions from the extreme wings of the strong rotational water vapor lines in the far infrared** as well as possible $(H_2O)_2$ dimer contributions (Ref. 11). Neither has been validated. However, it is important to note that either of these mechanisms would lead to the same pressure and temperature dependence. Therefore, the question of which mechanism is operative is mostly of fundamental interest.

We performed three separate analyses of the EMI data. The results of these show a consistent correlation with P_{H_2O} and integrated band transmittance in the 7.9-11.3 μm region and imply that the current LOWTRAN estimate of the continuum absorption coefficient over this bandwidth is too high and should be lowered by a factor of about 0.65 to 0.75. A similar analysis of the EMI 10.59 μm CO_2 laser transmission data implies a correction factor of 0.75 should be applied to the LOWTRAN value. The laser data at 10.59 μm , in fact, represents a cleaner evaluation of the water continuum, since at 10.59 μm there is very little contribution from H_2O molecular line absorption.

* In the 7.9-11.3 μm region, $\sigma_{H_2O \text{ mol}} \sim P_{H_2O}^{1/2}$ has a weaker water dependence than $\sigma_{H_2O \text{ cont}} \sim P_{H_2O}^2$ and is also usually smaller in magnitude.

** In our analyses and fit of the 8-12 μm window we will include continuum data out to 30 μm .

III. DISCUSSION OF RESULTS FROM THE EMI ANALYSIS

The EMI data, by itself, is probably insufficient evidence to warrant a change in the current LOWTRAN σ_{H_2O} cont. When we combined the EMI data with a review of other compatible values of σ_{H_2O} cont (*vide infra*) we became convinced of the validity of the correction factor.

By far the most useful data for our complete analysis are the CO_2 laser line measurements of Long and co-workers (Refs. 8, 9) together with the most recent data from Burch's laboratory (Ref. 12). Long's measurements,* based upon a White-type multipass optical system with an effective path length of 1 km, imply a correction to LOWTRAN σ_{H_2O} cont of 0.70 to 0.80, a value which is consistent with the EMI data. Very recent laser measurements by Aref'ev (Ref. 10) also support this value. This same range of correction factors are also consistent with a set of airborne IR radiance measurements in the 10-12 μm band reported by Platt (Ref. 13). Furthermore, we have performed a set of LOWTRAN calculations using the modified σ_{H_2O} cont on the recent transmission measurements made by Moulton (Ref. 5). There is a marked improvement in the match between the field measurements by Moulton and the computations based upon the modified σ_{H_2O} cont in the otherwise identical LOWTRAN 3 program. The Burch data, which spans the entire 8-12 μm region, is consistent with the above measurements at 10.59 μm .

* The most recent measurements from Long's group (Ref. 9) are essentially identical to the original measurements of McCoy et al. (Ref. 8) for the total H_2O continuum absorption at 760 torr ($H_2O + N_2$); however, the recent and more reliable results of Ref. 9 differ in the magnitude of the pure H_2O term. This paradox is resolved by assuming a smaller N_2 broadening coefficient (see discussion later in text).

Since the original Burch data was used in the LOWTRAN model, a paradox arises concerning the apparent consistency of Burch's values with the measurements cited above and the inconsistency obtained from the LOWTRAN value based on the same Burch data. We have resolved this problem by a closer inspection of the individual components that lead to the continuum absorption.

IV. T.E ABSORPTION MECHANISM

The two contributions are due to a self-broadening term $\sigma_{H_2O-H_2O}$ and an ambient one due to the collisions by other atmospheric gases (nominally N_2 , which comprises 79% of the total) written as $\sigma_{H_2O-N_2}$, so that

$$\sigma_{H_2O \text{ cont}} = \sigma_{H_2O-H_2O} + \sigma_{H_2O-N_2} . \quad (2)$$

In the current LOWTRAN routine the two terms are grouped together and expressed as

$$\sigma_{H_2O \text{ cont}} \approx C^0(v) w_{H_2O} \left[P_{H_2O} + \gamma \left(P - P_{H_2O} \right) \right] \quad (3)$$

where $C^0(v)$ is the self-broadening absorption coefficient as a function of frequency v measured at a temperature $T = 296K$, w_{H_2O} is the amount of water vapor in units of molecules cm^{-3} , P is the total atmospheric pressure in atmospheres, and γ is a relative measure of the ambient to self-broadened water continuum term. In the above units σ is then expressed in cm^{-1} . Aside from the assumption that $C^0(v)$ has the same shape for N_2-H_2O and H_2O-H_2O contributions, there are three possibilities for the introduction of errors in the above formulation; in particular, uncertainties in $C^0(v)$, γ , and, perhaps most critically, the lack or inclusion of an explicit temperature (T) dependence for $C^0(\dots)$. We will discuss each of these below.

V. ASSESSMENT OF N_2 - H_2O CONTRIBUTION

Of the three factors affecting the water vapor continuum, the greatest experimental uncertainty is with the N_2 - H_2O absorption. It is relatively small and therefore difficult to measure. Currently the LOWTRAN routine sets $\gamma = 0.005$, a value attributed to McCoy, Rensch, and Long (Ref. 8). This value was based upon a single measurement with a large experimental uncertainty. We believe 0.005 represents an upper bound to γ , since more recent measurements by Long's group (Ref. 9) suggest that this value is a substantial overestimate and that a more reasonable estimate is between 0 and 0.002. This is supported by Burch (Ref. 7), who was unable to detect *any* contribution to the H_2O continuum due to N_2 . If γ had been greater than about 0.001, Burch's experiments probably would have shown the effect of N_2 broadening. Burch has been able to measure $\sigma_{H_2O-N_2}$ in the neighboring 16-30 μm band, however (Ref. 14). In this region the relative contribution from H_2O-N_2 falls off rapidly with decreasing wavelength λ .* An extrapolation of Burch's γ values to 10 μm gives $\gamma \approx 0.0008$,** again much smaller than the value normally used in atmospheric modeling. Until better experimental measurements are made, the current LOWTRAN value for the N_2 - H_2O continuum should either be drastically reduced or preferably eliminated entirely since its magnitude, temperature

* In this paper we have used frequency ν (in cm^{-1}) and wavelength λ (in μm) interchangeably to define the spectral bands of interest:

$$\nu (cm^{-1}) = 10^4 / \lambda (\mu m).$$

** This value is based upon a linear regression using an exponential fit to 16 data points in the 16-30 μm range with $r^2 = 0.89$.

dependence, and frequency dependence are unknown in the 8-12 μm region. For conditions similar to the EMI measurements (4-14 torr water vapor pressure) the overall correction factor to the continuum coefficient induced by this change would be between 0.5 and 0.8, in total accord with the data analysis outlined above.

VI. DETERMINATION OF $C^0(v)$

We next address ourselves to improving the fit to $C^0(v)$ and the inclusion of a temperature dependence. Burch's original data, which was used for the LOWTRAN fit (Fig. 1 with open circles), exhibited a large amount of scatter near 8 μm , whereas his most recent values* do not. We have fit the new data by carrying out a linear regression in $\ln(C^0 - a)$ versus v . For all the fits presented below the best value of a was $1.25 \times 10^{-22} \text{ mol}^{-1} \text{ cm}^2 \text{ atm}^{-1}$. The absorption coefficient at $T = 296\text{K}$ is then given by

$$C^0(v) = a + b \exp(-\beta v). \quad (4)$$

This equation is not based upon any particular theoretical model but is rather meant to be a practical algorithm for calculating the water vapor continuum absorption coefficient. The experimental data and fits are shown in Figs. 1 and 2.

Figure 1 displays all available *laboratory* measurements for the water vapor continuum absorption at 296K. The open circles which are spuriously high (presumably due to possible impurities and perhaps some H_2O local molecular line contributions) near 8 μm represent the older Burch measurements. The solid circles are his most recent ones.* The solid triangles are recent CO_2 laser measurements by Long (Ref. 9) with a mixture of H_2O and N_2 at a total pressure of 760 torr. The water

* The original data measurements also included points from 10 to 12 μm ; however, these are coincident with the most recent measurements indicated as solid circles (●) in Fig. 1.

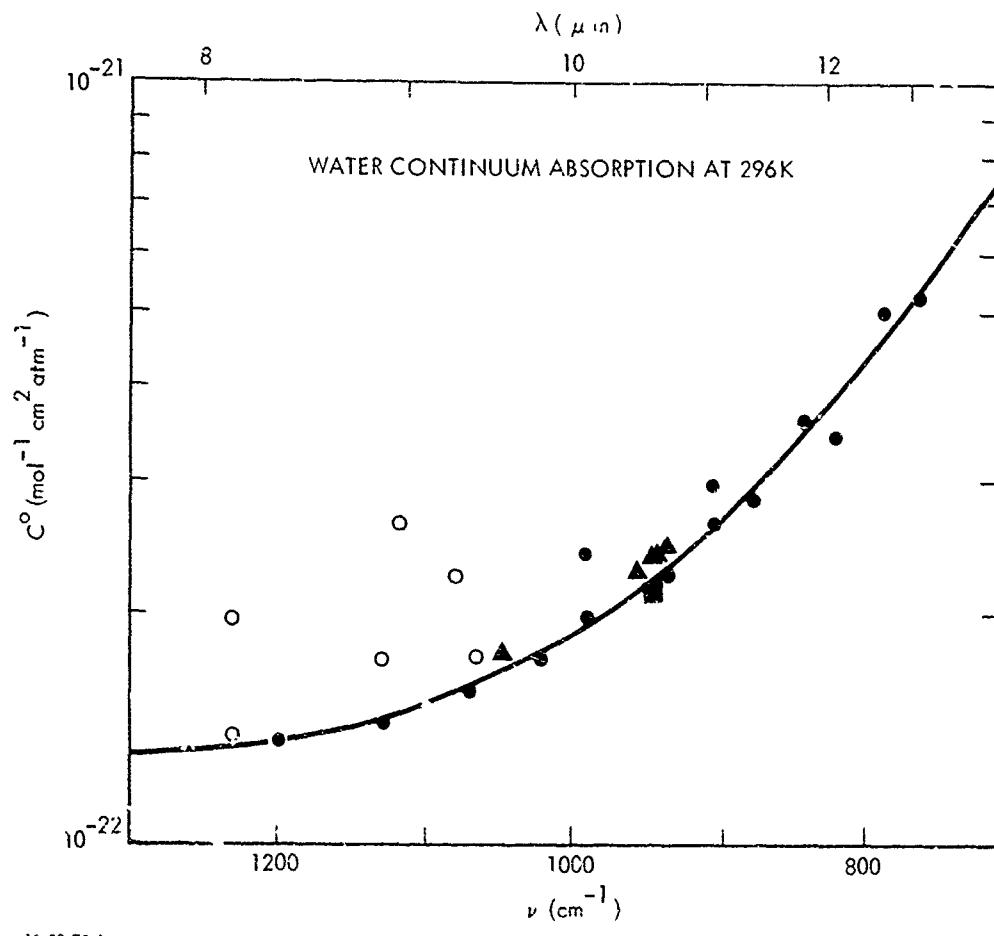


FIGURE 1. Water vapor continuum absorption coefficient $C(\nu)$ at $T = 296K$ as a function of frequency ν and wavelength λ in the $8-12 \mu\text{m}$ region; most recent data of Burch (Ref. 12) (\bullet); Original Burch Data (Ref. 7) (\circ); Mills and Long and Aref'ev data (Refs. 9 and 10) (\blacksquare); and Long measurements with N_2 and H_2O at a total pressure of 760 torr (Ref. 9) (\blacktriangle). The solid line represents a linear regression (described in text) to all of Burch's recent water vapor data for the $8-30 \mu\text{m}$ Region.

vapor pressure was 14.3 torr at a temperature of 294K corresponding to mid-latitude summer conditions.* Although these results are somewhat higher than for pure H_2O , the cumulative error of all of the data shown precludes the possibility of extracting anything but a crude estimate of the N_2 - H_2O broadening term. The solid square is the 10.59 μm measurement of Mills and Long (Ref. 9) and Aref'ev (Ref. 10). The solid line represents a linear regression using Eq. 4 on all of the recent Burch values, including those made out to 30 μm . The coefficient of determination (r^2) for this fit is $r^2 = 0.992$ for 36 data points. The values of the fit parameters used to generate this line are

$$a = 1.25 \times 10^{-22} \text{ mol}^{-1} \text{ cm}^2 \text{ atm}^{-1},$$

$$b = 1.67 \times 10^{-19} \text{ mol}^{-1} \text{ cm}^2 \text{ atm}^{-1},$$

and

$$\beta = 7.87 \times 10^{-3} \text{ cm}.$$

In Fig. 2 all of the recent Burch data (solid circles) is shown together with the above fit, and a dashed curve that represents a linear regression to the Burch data** in the 8-12 μm vicinity but with $r^2 = 0.994$. This fit extrapolates very well into the 30 μm range, and is also virtually indistinguishable from the previous fit for the 8-12 μm window. The fit parameters are given by

$$a = 1.25 \times 10^{-22} \text{ mol}^{-1} \text{ cm}^2 \text{ atm}^{-1},$$

$$b = 2.34 \times 10^{-19} \text{ mol}^{-1} \text{ cm}^2 \text{ atm}^{-1},$$

and

$$\beta = 8.30 \times 10^{-3} \text{ cm}.$$

Some of the scatter shown in Fig. 2 for the 12-30 μm region is due to the contribution by neighboring molecular lines.

* Mid-latitude summer conditions as typified by the LOWTRAN model (Ref. 4).

** For this restricted linear regression we have eliminated the anomalously high values (probably due to small local line contributions or impurities) whenever two measurements were made at nearly the same ν (i.e., at 990, 904, 844, and 787 cm^{-1}).

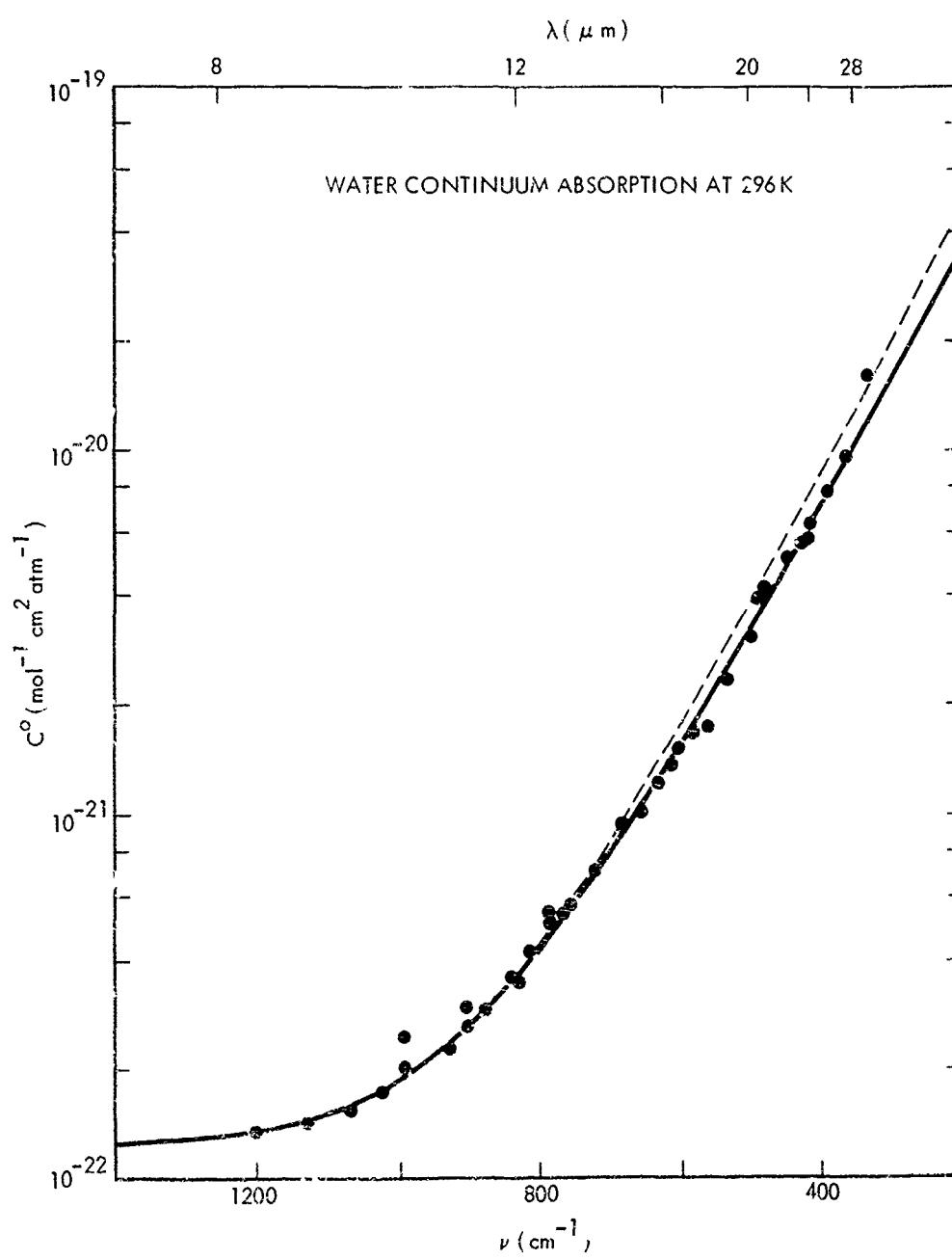


FIGURE 2. Water vapor continuum absorption coefficient $C(v)$ at $T = 296K$ as a function of frequency v and wavelength λ in the $8-30 \mu\text{m}$ region; most recent data of Burch (Refs. 12 and 14) (\bullet). The solid line represents a linear regression (described in text) to all of Burch's recent water vapor data for the $8-30 \mu\text{m}$ Region. The dashed curve represents a linear regression to the best Burch data in the $8-12 \mu\text{m}$ region (described in text).

Presumably, if one subtracted the local line values from the experimental ones, a much smoother curve would result. The current data therefore represents an upper bound to the true water vapor continuum. The above fits are nearly the same as the current LOWTRAN versions near 12 μm but are about 20-25% lower near 8 μm . It is noteworthy that the EMI 7.9-11.3 μm measurements are much more sensitive to the extinction coefficient at the 8 μm end of the spectrum.* We next address the problem of the temperature dependence for the water vapor continuum.

*The broadband EMI transmission measurements were made with a carbon arc light source with a blackbody temperature of about 3800K. Hence, the photon density near 8 μm is substantially larger than at 12 μm .

VII. TEMPERATURE DEPENDENCE

The final and most critical change to be made for the LOWTRAN water continuum model is the inclusion of a strong measured temperature dependence in the 8-12 μm window. Although the temperature dependence for the water vapor continuum absorption had been recognized before, it was not utilized in the atmospheric transmission models. The collected data of Burch (Ref. 7), Aref'ev et al. (Ref. 10), Bignell (Ref. 15), and Varanasi et al. (Ref. 11), together with an absorption model based upon dimer contributions (Ref. 15), suggests

$$C(v, T) = C^0(v) \exp \left[\frac{T_0}{T} - \frac{1}{296} \right], \quad (5)$$

where $C^0(v)$ is the absorption coefficient at 296K. Although the fit for $C^0(v)$ is accurate beyond the 8-12 μm region, the above temperature dependence breaks down (if a single v -independent T_0 is used) near 20 μm , where the temperature dependence is much weaker. Also, beyond the 8-12 μm vicinity the $\text{N}_2\text{O}-\text{N}_2$ broadening becomes much more important. Thus one should be wary of an application of Eq. 5 outside the 8-12 μm region for temperatures other than ca. 296K. The mean representative values of T_0 are given in Table 1. Although it is not shown in Table 1, it is interesting to note that an extrapolation of Burch's 12-30 μm data also yields $T_0 \approx 1800\text{K}$. Varanasi et al. (Ref. 11) first suggested that dimers might be responsible for the strong temperature dependence of $C(v, T)$. This is in accord with the fact that the hydrogen bond between two water monomers is in the neighborhood of 3-4 kcal/mole (Ref. 16), which leads to $T_0 \approx 1800\text{K}$. In fact, any reasonable line shape theory

incorporating the strongly attractive interaction between two water molecules during their collision will lead to a similar dependence.

TABLE I. TEMPERATURE DEPENDENCE PARAMETER T_0 FOR THE 8-12 μm PURE WATER VAPOR CONTINUUM ABSORPTION COEFFICIENT ACCORDING TO

$$C(v, T) = C^0(v) \exp \left[T_0 \left(\frac{1}{T} - \frac{1}{296} \right) \right]^*$$

<u>REFERENCE</u>	<u>T_0 (K)</u> **
Burch (Ref. 7)	1745
Aref'ev (Ref. 10)	1810
Bignell (Ref. 15)	1800
Varanasi et al. (Ref. 11)	2000
Dimer Model (Refs. 15,16)	1750
Best Estimate	1800

* $C^0(v)$ is the water vapor absorption coefficient at $T = 296\text{K}$.

** The values for T_0 cited are simply mean representative ones. The range of uncertainty can be as high as $\pm 500\text{K}$ for a given measurement (i.e., Varanasi et al.). A more typical range is $\pm 200\text{K}$.

VIII. SUMMARY

In the foregoing discussion we have suggested three modifications to the current LOWTRAN atmospheric model for water vapor continuum absorption in the 8-12 μm infrared window region:

1. Improved fit to $C^0(v)$ that is about 25% lower than the current model at 8 μm but nearly the same at 12 μm
2. Elimination or reduction of the uncertain $\text{N}_2\text{-H}_2\text{O}$ broadening term
3. Most critically, addition of a strong temperature dependence to the H_2O continuum absorption.

A consequence of the first two improvements is an increase of the computed atmospheric transmission for 10, 20, and 50 km path lengths and 85% relative humidity at 296K by factors of ca. 2, 5, and 53, respectively, for 10.59 μm radiation. Furthermore, since diurnal temperature changes of 15°C are not uncommon, the temperature factor alone can lead to daily variations by factors of ca. 3, 6, and 90, respectively, for the same wavelength and total amount of H_2O vapor. On a monthly or seasonal basis, where variations of 30°C or more are typical, the modification is by factors of ca. 7, 50, and greater than 10,000! For certain applications, especially for a long path at ground level, spatial and temporal variations of temperature and water vapor pressure along the path can be important and should be accounted for in applying the model presented here.

In summary, for high-visibility conditions where aerosols are relatively unimportant, one can now expect to do meaningful

analyses and design of infrared equipment and have measured performance closer to the computed expectations. The problems remaining in the area of water vapor absorption are an improved measurement of the N_2 - H_2O contribution in the 8-12 μm region, an inclusion of the H_2O continuum to the 3-5 μm vicinity, and the development of a viable theoretical model for the continuum line shapes and temperature profiles.

The model for the 8-14 μm water vapor continuum presented here is currently being incorporated into the LOWTRAN 3 model together with a 3-5 μm water vapor continuum model. The next version of the LOWTRAN code will also contain additional aerosol models for maritime, rural, and urban atmospheres.

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